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U.S. DEPARTMENT OF COMMERCE
PATENT AND TRADEMARK OFFICE

ATTORNEY'S DOCKET NUMBER
44599.

TRANSMITTAL LETTER TO THE UNITED STATES DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. 371

U.S. APPLICATION NO. (if known, see 37 C.F.R. 1.5)
10/069193

INTERNATIONAL APPLICATION NO.	INTERNATIONAL FILING DATE	PRIORITY DATE CLAIMED
PCT/US00/24915	**08 September 2000**	**23 September 1999**

TITLE OF INVENTION
SOLVENT COMPOSITION

APPLICANT(S) FOR DO/EO/US
Martin F. Hill; Rene R. Blank; Alan Goodson; Manuela Ehreiser

Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:

1. [X] This is a FIRST submission of items concerning a filing under 35 U.S.C. 371.
2. [ ] This is a SECOND or SUBSEQUENT submission of items concerning a filing under 35 U.S.C. 371.
3. [ ] This is an express request to begin national examination procedures (35 U.S.C. 371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b) and PCT Articles 22 and 39(1).
4. [X] A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date.
5. [X] A copy of the International Application as filed (35 U.S.C. 371(c)(2))
   a. [ ] is transmitted herewith (required only if not transmitted by the International Bureau).
   b. [ ] has been transmitted by the International Bureau.
   c. [X] is not required, as the application was filed in the United States receiving Office (RO/US).
6. [ ] A translation of the International Application into English (35 U.S.C 371(c)(2)).
7. [X] Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3))
   a. [ ] are transmitted herewith (required only if not transmitted by the International Bureau).
   b. [ ] have been transmitted by the International Bureau.
   c. [ ] have not been made; however, the time limit for making such amendments has NOT expired.
   d. [X] have not been made and will not be made.
8. [ ] A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).
9. [X] An oath or declaration of the inventor (35 U.S.C. 371(c)(4)).
10. [ ] A translation of the Annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)).

Items 11. to 15. below concern other document(s) or information included:

11. [X] An Information Disclosure Statement under 37 CFR 1.97 and 1.98.
12. [X] A FIRST preliminary amendment.
    [ ] A SECOND or SUBSEQUENT preliminary amendment.
13. [ ] A substitute specification.
14. [ ] A change of power of attorney and/or address letter.
15. [ ] Other items or information:

U.S. APPLICATION NO. (If known, see 37 C F.R 1.50)	INTERNATIONAL APPLICATION NO.	ATTORNEY'S DOCKET NUMBER
10/069193	PCT/US00/24915	44599

17. [X] The following fees are submitted:	CALCULATIONS	PTO USE ONLY
Basic National Fee (37 CFR 1.492(a)(1)-(5)):		
Search Report has been prepared by the EPO or JPO . . . . $ 890.00		
International preliminary examination fee paid to USPTO (37 CFR 1.482) . . . . . $ 710.00		
No international preliminary examination fee paid to USPTO (37 CFR 1.482) but international search fee paid to USPTO (37 CFR 1.445(a)(2)) . . . . . $ 740.00		
Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445 (a)(2)) paid to USPTO . . . . . $ 1,040.00		
International preliminary examination fee paid to USPTO (37 CFR 1.482) and all claims satisfied provisions of PCT Article 33(2)-(4) . . . . . $ 100.00		
ENTER APPROPRIATE BASIC FEE AMOUNT =	$ 890.00	
Surcharge of $ 0.00 for furnishing the oath or declaration later than [ ] 20 [ ] 30 months from the earliest claimed priority date (37 CFR 1 492(e)). +	$ 0.00	

Claims	Number Filed	Number Extra	Rate		
Total Claim	12 - 20 =	0	X $ 18.00	$ 0.00	
Independent Claims	4 - 3 =	1	X $ 84.00	$ 84.00	
Multiple dependent claim(s) (if applicable)			$ 0.00	$ 0.00	
Processing fee of $ 0 for furnishing the English Translation later than [ ] 20 [ ] 30 months from the earliest claimed priority date (37 CFR 1.492(f)). +				$ 0.00	
TOTAL NATIONAL FEE =				$ 974.00	
				Amount to be refunded:	$
				charged:	$

a. [ ] A check in the amount of $_____ to cover the above fees is enclosed.

b. [X] Please charge my Deposit Account No. **04-1512** in the amount of $ **974.00** to cover the above fees. A duplicate copy of this sheet is enclosed.

c. [X] The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No. **04-1512** . A duplicate copy of this sheet is enclosed.

Note: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137(a) or (b)) must be filed and granted to restore the application to pending status.

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Date: **14 February 2002**

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicant (s): Hill et al.

Attorney Docket No.: 44599

Filed: Concurrently Herewith

For: SOLVENT COMPOSITION

"Express Mail" mailing number **EL636831253US**
Date of Deposit **February 14, 2002**

Assistant Commissioner of Patents
Washington, D.C. 20231

Sir:

PRELIMINARY AMENDMENT

Please amend Claims 4-7 and 10 as follows, cancel Claim 12 and add the following Claims 13-21.

4. (Amended) The solvent composition of Claim 1 wherein component b) is butylene carbonate, propylene carbonate or ethylene carbonate.

5. (Amended) The solvent composition of Claim 1 wherein component c) is water.

6. (Amended) The solvent composition of Claim 1 comprising a) a propylene glycol monoether, b) propylene carbonate and c) water.

7. (Amended) The solvent composition of Claim 1 comprising a) a propylene glycol monoether and b) ethylene carbonate.

10. (Amended) The composition of Claim 8 wherein the solvent composition comprises a) at least 50 percent of a glycol monoether or diether, b) from 1 to 25 percent of an organic low molecular weight compound containing a carbonate group and c) from 0 to 25 percent of a protic liquid other than a glycol monoether or diether, based on the total weight of a), b) and c).

Claim 12 has been cancelled.

13. The solvent composition of Claim 2 wherein component b) is butylene carbonate, propylene carbonate or ethylene carbonate.

14. The solvent composition of Claim 3 wherein component b) is butylene carbonate, propylene carbonate or ethylene carbonate.

15. The solvent composition of Claim 2 wherein component c) is water.

16. The composition of Claim 8 wherein the solvent composition comprises a) from 60 to 90 percent of a glycol monoether, b) from 5 to 15 percent of an organic low molecular weight compound containing a carbonate group and c) from 0 to 25 percent of a protic liquid other than a glycol monoether or diether, based on the total weight of a), b) and c).

17. The composition of Claim 8 wherein component a) in the solvent composition is a propylene or butylene glycol monoether represented by Formula I

$$R^1O - (CH_2 - CHR^3O)_n - R^2 \quad (I)$$

wherein

one of the substituents R^1 and R^2 is an alkyl group having from 1 to 12 carbon atoms and the other of the substituents R^1 and R^2 is hydrogen,

R^3 in each occurrence independently is methyl or ethyl and

n is from 1 to 4.

18. The composition of Claim 8 wherein component b) in the solvent composition is butylene carbonate, propylene carbonate or ethylene carbonate.

19. The composition of Claim 8 wherein component c) in the solvent composition is water.

20. The composition of Claim 8 wherein the solvent composition comprises a) a propylene glycol monoether, b) propylene carbonate and c) water.

21. The composition of Claim 8 wherein the solvent composition comprises a) a propylene glycol monoether and b) ethylene carbonate.

Remarks

The amendments in Claims 4-7 only relate to formal matters. The amendment in Claim 10 is based on page 3, paragraph 1 of the original specification. New Claims 13 and 14 are based on original Claim 4. New Claim 15 is based on original Claim 5.

New Claims 16-21 are based on original Claim 10 in combination with original Claims 1-7.

Respectfully submitted,

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VERSION WITH MARKINGS TO SHOW CHANGES MADE

Claim 12 has been cancelled. Claims 4-7 and 10 have been amended and Claims 13-21 have been added.

4. (Amended) The solvent composition of ~~any one of Claims 1 to 3~~ Claim 1 wherein component b) is butylene carbonate, propylene carbonate or ethylene carbonate.

5. (Amended) The solvent composition of ~~any one of Claims 1 to 4~~ Claim 1 wherein component c) is water.

6. (Amended) The solvent composition of ~~any one of Claims 1 to 5~~ Claim 1 comprising a) a propylene glycol monoether, b) propylene carbonate and c) water.

7. (Amended) The solvent composition of ~~any one of Claims 1 to 5~~ Claim 1 comprising a) a propylene glycol monoether and b) ethylene carbonate.

10. (Amended) The composition of Claim 8 ~~comprising a solvent composition set forth in any one of claims 1 to 7.~~ wherein the solvent composition comprises a) at least 50 percent of a glycol monoether or diether, b) from 1 to 25 percent of an organic low molecular weight compound containing a carbonate group and c) from 0 to 25 percent of a protic liquid other than a glycol monoether or diether, based on the total weight of a), b) and c).

Claim 12 has been cancelled.

13. The solvent composition of Claim 2 wherein component b) is butylene carbonate, propylene carbonate or ethylene carbonate.

14. The solvent composition of Claim 3 wherein component b) is butylene carbonate, propylene carbonate or ethylene carbonate.

15. The solvent composition of Claim 2 wherein component c) is water.

16. The composition of Claim 8 wherein the solvent composition comprises a) from 60 to 90 percent of a glycol monoether, b) from 5 to 15 percent of an organic low molecular weight compound containing a carbonate group and c) from 0 to 25 percent of a protic liquid other than a glycol monoether or diether, based on the total weight of a), b) and c).

17. The composition of Claim 8 wherein component a) in the solvent composition is a propylene or butylene glycol monoether represented by Formula I

$$R^1O - (CH_2 - CHR^3O)_n - R^2 \quad (I)$$

wherein

one of the substituents R^1 and R^2 is an alkyl group having from 1 to 12 carbon atoms and the other of the substituents R^1 and R^2 is hydrogen,

R^3 in each occurrence independently is methyl or ethyl and

n is from 1 to 4.

18. The composition of Claim 8 wherein component b) in the solvent composition is butylene carbonate, propylene carbonate or ethylene carbonate.

19. The composition of Claim 8 wherein component c) in the solvent composition is water.

20. The composition of Claim 8 wherein the solvent composition comprises a) a propylene glycol monoether, b) propylene carbonate and c) water.

21. The composition of Claim 8 wherein the solvent composition comprises a) a propylene glycol monoether and b) ethylene carbonate.

10/069193

SOLVENT COMPOSITION

The present invention relates to a solvent composition and to the use of the solvent composition as a diluent for one or more epoxy resins and/or one or more curing agents for epoxy resins and/or one or more curing catalysts and/or one or more cure inhibitors.

Background of the Invention

It is a well known technique to utilize an organic solvent for reducing the viscosity of liquid epoxy resins or liquid curing agents therefor or to solubilize solid resins and/or curing agents in organic solvents in order to facilitate the handling of the epoxy resins and/or of the curing agents.

Excellent solvents for curing agents for epoxy resins, such as dicyandiamide, are dimethyl formamide and methyl glycol. However, the use of dimethyl formamide or methyl glycol has raised industrial hygiene concerns.

Accordingly, one object of the present invention is to find new solvents which are useful for dissolving epoxy resins, curing agents for epoxy resins, curing catalysts or cure inhibitors or which are useful for reducing the viscosity of such compounds.

Known uses of epoxy resins are among others the production of electrical laminates, glass laminates and coatings. In a typical procedure a solution of an epoxy resin in an organic solvent, such as a ketone, and a solution of a curing agent for an epoxy resin, such as dicyandiamide, are combined to produce an epoxy resin composition which is used in various applications. When using known solvents for dicyandiamide, such as propylene glycol monomethyl ether, often difficulties have been experienced in keeping dicyandiamide in solution after the dicyandiamide solution has been mixed with the epoxy resin solution.

Accordingly, a preferred object of the present invention is to find new solvents in which curing agents for an epoxy resin,

such as dicyandiamide, are sufficiently soluble even after the curing agent solution has been mixed with an epoxy resin solution.

Summary of the Invention

One aspect of the present invention is a solvent composition which comprises:

a) a glycol monoether or diether and

b) an organic low molecular weight compound containing a carbonate group.

Another aspect of the present invention is a composition which comprises one or more compounds selected from i) epoxy resins, ii) curing agents for an epoxy resin, iii) curing catalysts, and iv) cure inhibitors in a solvent composition indicated above.

Yet another aspect of the present invention is a method of preparing a composition comprising one or more compounds selected from i) epoxy resins, ii) curing agents for an epoxy resin, iii) curing catalysts, and iv) cure inhibitors, in a solvent composition by contacting one or more such compounds with

a) a glycol monoether or diether and

b) an organic low molecular weight compound containing a carbonate group.

Yet another aspect of the present invention is the use of a) a glycol monoether or diether and b) an organic low molecular weight compound containing a carbonate group as a diluent for one or more compounds selected from i) epoxy resins, ii) curing agents for an epoxy resin, iii) curing catalysts, and iv) cure inhibitors.

Detailed Description of the Invention

The solvent composition of the present invention preferably comprises

a) at least 45 percent, preferably at least 50 percent, more preferably at least 60 percent, and most preferably from 60 to 90 percent, of a glycol monoether or diether; and

b) from 1 to 30 percent, preferably from 1 to 25 percent, more preferably from 1 to 20 percent, most preferably from 5 to 15 percent, of an organic low molecular weight compound containing a carbonate group and; optionally

c) up to 25 percent, preferably up to 20 percent, more preferably from 2 to 15 percent, most preferably from 5 to 12 percent, of a protic liquid other than a glycol monoether or diether, based on the total weight of a), b) and c).

Component a) of the solvent composition is a glycol monoether or a glycol diether or a mixture of one or more monoethers and/or diethers. The monoethers are preferred over the diethers.

Preferred glycol monoethers are the propylene or butylene glycol monoethers, most preferably ethers represented by Formula I

$$R_1O - (CH_2 - CHR^3O)_n - R_2 \qquad (I)$$

wherein

one of the substituents R_1 and R_2 is an alkyl group having from 1 to 12, preferably from 1 to 6, more preferably from 1 to 4 carbon atoms and the other of the substituents R_1 and R_2 is hydrogen,

R^3 in each occurrence independently is ethyl or, preferably, methyl and

n is from 1 to 4, preferably 1, 2 or 3.

The alkyl groups may be branched or unbranched. Exemplary of the alkyl groups are methyl, ethyl, n-propyl, isopropyl, the butyl groups, such as n-butyl or isobutyl, and the pentyl, hexyl, octyl, decyl or dodecyl groups. Of the propyl and butyl groups n-propyl and n-butyl are preferred. Preferably, one of the substituents R_1 and R_2 is methyl or n-butyl.

Preferred monoethers of Formula I are propylene glycol n-butyl ether, dipropylene glycol methyl ether, dipropylene glycol n-butyl ether and, most preferably, propylene glycol methyl ether.

The corresponding propylene or butylene glycol diethers wherein both substituents R_1 and R_2 are an alkyl group are also useful.

Component b) of the solvent composition is an organic low molecular weight compound containing a carbonate group. By the term "low molecular weight" as used herein is meant a molecular weight of up to 200, preferably of up to 150, most preferably up to 120. A useful component b) is for example ethylene carbonate or butylene carbonate. The preferred component b) is propylene carbonate.

The optional component c) of the solvent composition is a protic liquid, other than a glycol monoether or diether, or a mixture of two or more such protic liquids. By "liquid" is meant a compound that is liquid at room temperature and atmospheric pressure. The most preferred protic liquid is water. Other protic liquids are alcohols or glycols, such as methanol, ethanol, ethylene glycol or propylene glycol.

If component b) of the solvent composition is propylene carbonate or butylene carbonate, it is advisable to include component c), preferably water, in the solvent composition of the present invention. If component b) of the solvent composition is ethylene carbonate, the solvent composition is an excellent solvent for a curing agent, such as dicyandiamide, with or without component c).

Depending on the intended use of the solvent composition of the present invention, it preferably contains one or more other solvents in addition to components a), b) and c), preferably ketones, such as acetone, methyl ethyl ketone or methyl isobutyl ketone; or amides, such as dimethyl formamide. Their amount also depends on the intended use of the solvent composition.

If the solvent composition of the present invention is used for diluting an epoxy resin, the total amount of the components

a), b) and c) generally amounts to at least 40 percent, preferably at least 50 percent, more preferably at least 60 percent and the amount of one or more other solvents is generally up to 60 percent, preferably up to 50 percent, more preferably up to 40 percent, based on the total weight of the solvents in the solvent composition.

If the solvent composition is used as a diluent for a curing agent for an epoxy resin, a curing catalyst or a cure inhibitor, the total amount of the components a), b) and c) generally amounts to at least 60 percent, preferably at least 75 percent, more preferably at least 90 percent and the amount of one or more other solvents is generally up to 40 percent, preferably up to 25 percent, more preferably up to 10 percent, based on the total weight of the solvents in the solvent composition.

Most preferably, the solvent composition of the present invention substantially consists of the components a), b) and c).

The solvent composition may comprise other additives, such as viscosity modifiers, for example N-methyl pyrrolidone, thickeners, for example high molecular weight polyalkylene glycols, or plasticizers, for example dioctyl phthalate or chlorinated paraffin. If present, their amount preferably is from 0.1 to 20 percent, more preferably from 1 to 10 percent, based on the total weight of components a), b) and c).

The solvent composition of the present invention may comprise solid particles, such as a filler. However, the solvent composition does not comprise more than 150 percent, preferably not more than 100 percent of solid particles, based on the total weight of components a), b) and c). Useful fillers include organic and inorganic fillers, such as melamin resins, wood fillers, carbon black or graphite, talc, calcium carbonate, phosphates, such as ammonium polyphosphate, flyash, aluminium trihydroxide, magnesium hydroxide, glass fibers, marble dust, cement dust, clay, feldspar, silica or glass, fumed silica, alumina, magnesium oxide, zinc oxide, barium sulfate, aluminum silicate, calcium silicate, titanium dioxide, titanates, glass microspheres or chalk. Useful fire retardant fillers and additives, such as sulfur compounds,

phosphorus compounds, boron compounds, silicon compounds and polynuclear aromatic compounds are for example described in "International Plastics Flammability Handbook" by Jürgen Troitzach, 1983, ISBN 0-02-949770-1 Macmillan Publishing Co., Inc., New York pages 46 to 53 and "Ullmann's Encyclopedia of Industrial Chemistry" Vol. A11 pages 124 to 126.

The solvent composition of the present invention can be produced in a known manner. Generally the components of the solvent composition are mixed in the ratios indicated above at a temperature of from 1°C to 80°C, preferably from 15°C to 40°C, until a homogeneous mixture is obtained.

The solvent composition of the present invention is very useful as a diluent for one or more compounds selected from i) epoxy resins, ii) curing agents for an epoxy resin, iii) curing catalysts, and iv) cure inhibitors. The diluent can act as a means for reducing the viscosity of one or more liquid compounds or as a solvent.

Preferably, a curing agent for an epoxy resin, optionally a curing catalyst and optionally a cure inhibitor is diluted with the solvent composition of the present invention. This blend is preferably mixed with an epoxy resin which is diluted with an organic solvent.

Curing agents for epoxy resins, commonly also called epoxy hardeners, are well known in the art. Useful classes of curing agents are for example amides, acid anhydrides, such as styrene/maleic anhydrides, boron trifluoride complexes, dicyandiamide, substituted dicyandiamides, polyester resins, novolacs or phenolic hardeners, that is compounds containing more than one aromatic hydroxyl group. Another class of curing agents well known in the art comprises prereacted adducts of epoxy resins with amines or anhydrides or dicyandiamide or phenolic resins. Preferred phenolic hardeners are described on pages 6 to 8 of European patent specification 0,240,565. Other known curing agents are primary or secondary amines, hydrazides or hydrazine, preferably the multifunctional, more preferably the di- to hexafunctional

primary amines, amides and hydrazides. Such curing agents are listed in column 5, lines 47 to 68 and column 6, lines 14 to 19 of U.S. Patent 4,789,690.

Further useful curing agents are listed on page 11, lines 41-58 and page 12, lines 1-40 of the published European patent application EP-A-0,458,502. Other preferred curing agents are derivatives of cyanamide or dicyanamide, dihydroxy phenols, biphenols, halogenated bisphenols, alkylated bisphenols, trisphenols, phenol-aldehyde resins, halogenated phenol-aldehyde novolac resins, alkylated phenol-aldehyde novolac resins, hydrocarbon-phenol resins, hydrocarbon-halogenated phenol resins, hydrocarbon-alkylated phenol resins or a combination of two or more thereof.

The solvent composition of the present invention is particularly useful for dissolving a dicyandiamide, such as a substituted dicyandiamide or non-substituted dicyandiamide (cyanoguanidine). The solvent composition of the present invention is also useful for dissolving substituted dicyandiamides, such as dicyandiamides wherein some, but not all, hydrogens bonded to a nitrogen are replaced by alkyl, preferably C_{1-6}-alkyl, more preferably methyl, ethyl or the propyl groups; or by aryl, preferably benzyl, more preferably 2-methylbenzyl. Preferably, the dicyandiamide carries only one of the above-listed substituents. Most preferably, the dicyandiamide is not substituted.

The solvent composition of the present invention is useful for dissolving one or more of the above-mentioned curing agents for epoxy resins. The term "a curing agent" as used herein also comprises mixtures of two or more compounds which act as a curing agent for an epoxy resin.

Preferably from 1 to 20 percent, more preferably from 2 to 15 percent, most preferably from 3 to 12 percent of a curing agent is dissolved in the solvent composition, by the weight of the solvent composition. It is to be understood that the solubility of the curing agent in the solvent composition of the present invention depends on various factors, such as the type of the curing agent,

the specific composition of the solvent composition and the amounts and types of compounds which may additionally be dissolved in the solvent composition, such as a curing catalyst or a cure inhibitor. The solubility of a specific curing agent in a specific solvent composition of the present invention can be evaluated by series trials.

Curing catalysts or curing accelerators which increase the speed of reaction between the curing agent and the epoxy resin are also well known in the art. Preferred are tertiary amine-containing or heterocyclic amine containing compounds. Some curing catalysts may have an effect as a curing agent per se, such as benzoguanamidine, imidazoles, benzodimethylamine, metaphenolene diamine, or N,N,N',N'-tetramethyl-1,3-butadiamine. Preferred imidazoles are 2-methyl imidazole, 2-ethyl-4-methyl-imidazole or 2-phenylimidazole. 2-Methyl imidazole, 2-ethyl-4-methylimidazole and 2-phenylimidazole are the most preferred curing catalysts. Other curing catalysts which may be dissolved in the solvent composition of the present invention are heterocyclic nitrogen compounds, phosphines, sulfides or ammonium, phosphonium or sulfonium containing compounds. Such curing catalysts are listed on page 12, lines 41 to 50, on pages 13 to 16 and on page 17, lines 1 to 22 of the published European patent application EP-A-0,458,502. The solvent composition of the present invention is useful for dissolving one or more of the above-mentioned curing catalysts. The term "a curing catalyst" as used herein also comprises mixtures of two or more compounds which influence the speed of reaction between an epoxy resin and an epoxy hardener.

If the solvent composition is used for dissolving a curing catalyst, generally from 0.1 to 40 percent, preferably from 0.5 to 35 percent, more preferably from 1 to 20 percent and most preferably from 2 to 12 percent of a curing catalyst is dissolved in the solvent composition, by the weight of the solvent composition. It is to be understood that the solubility of the curing catalyst in the solvent composition of the present invention depends on various factors, such as the type of curing catalyst, the amount and type of curing agent which is optionally present in the solvent composition,

the specific composition of the solvent composition and the amounts and types of compounds which may additionally be dissolved in the solvent composition. The solubility of a specific curing catalyst in a specific solvent composition of the present invention can be evaluated by series trials. In the case of imidazoles such as 2-methylimidazole, preferably from 1 to 20, more preferably from 2 to 12 percent of an imidazole is dissolved in the solvent composition, by the weight of the solvent composition.

Useful cure inhibitors are boric acid, metaboric acid, boric acid anhydride or maleic acid or a mixture of (meta)boric acid (anhydride) with at least one acid having a weak nucleophilic anion, such as fluoroboric acid (HBF_4). Cure inhibitors are described on page 17, lines 18 to 50 and page 18, lines 1 to 38 of the published European patent application EP-A-0,458,502. If a cure inhibitor is dissolved in the solvent composition, it is preferably dissolved in the solvent composition in an amount of from 0.1 to 12 percent, more preferably from 0.5 to 6 percent, most preferably of from 1 to 4 percent , by the weight of the solvent composition. If the solvent composition of the present invention comprises substantial amounts of a curing agent, for example between 3 and 12 percent of dicyandiamide, the solubility of the curing catalyst and/or the cure inhibitor in the solvent composition is generally smaller, usually between 0.1 and 3 percent, typically between 0.5 and 2 percent, based on the weight of the solvent composition.

The desired amount of curing agent(s) and/or curing catalyst(s) and/or cure inhibitor(s) is added to the solvent composition and the resulting mixture is preferably stirred until a clear solution is obtained. The compounds to be dissolved can be added together or alternatingly to the solvent composition of the present invention. Alternatively, each compound can be dissolved individually in the solvent composition and the resulting solutions can be combined if desired. Preferred compositions of the resulting solutions of the present invention are described above.

According to another aspect of the invention, one or more compounds selected from curing agents for an epoxy resin, curing catalysts, and cure inhibitors are first dissolved in component a)

or component b) and optionally another solvent of the solvent composition and component b) or a) is subsequently added. For example, dicyandiamide is preferably first dissolved in a glycol monoether or diether a) and the optional component c) and at a later stage component b) is added.

Another preferred aspect of the present invention is an epoxy resin composition comprising an epoxy resin and preferably one or more compounds selected from curing agents for an epoxy resin, curing catalysts and cure inhibitors, in a solvent composition described further above.

The epoxy resin composition can be prepared in a known manner. According to a preferred method, an above-described solution of a curing agent and/or a curing catalyst and/or a cure inhibitor in the solvent composition of the present invention is mixed with an epoxy resin. Typically the epoxy resin is diluted with a solvent. Although the solvent for the epoxy resin can be added simultaneously or after the epoxy resin has been mixed with the solution of the curing agent, curing catalyst and/or cure inhibitor, the epoxy resin is preferably pre-mixed with the solvent. The epoxy resin solution is then mixed with the solution of the curing agent, curing catalyst and/or cure inhibitor for producing an epoxy resin composition which is typically designated in the art as "one-component epoxy resin composition".

The epoxy resin composition may comprise a wide variety of epoxy resins, provided that they are curable, preferably with dicyandiamide. Curable epoxy resins are well known in the art. Illustrative examples of epoxy resins useful herein are described in The Handbook of Epoxy Resins by H. Lee and K. Neville, published in 1967 by McGraw-Hill, New York, in appendix 4-1, pp. 4 through 56, and U.S. Patent Nos. 2,633,458; 3,477,990; 3,821,243; 3,970,719; 3,975,397; 4,071,477; and 4,582,892, and GB Patent Specification No. 1,597,610.

Useful solvents or diluents for the epoxy resin are well known in the art. Preferred examples are 2-methyl-pentanediol-(2,4), toluene, o-dichlorobenzene, cyclohexanone, cyclohexanol or,

more preferably, a ketone, such as acetone, methyl ethyl ketone or methyl isobutyl ketone. Mixtures of different solvents are also useful for dissolving the epoxy resin. Preferably, the epoxy resin is dissolved in one, two, three or more of the components of the solvent composition of the present invention. Generally, from 50 to 95 percent, preferably from 60 to 90 percent, more preferably from 70 to 85 percent epoxy resin is dissolved in or diluted with a suitable solvent, based on the total weight of epoxy resin and solvent.

The epoxy resin composition preferably comprises from 0.05 to 10 percent, more preferably from 0.1 to 6 percent, most preferably from 0.5 to 4 percent of a curing agent, such as dicyandiamide, based on the weight of the epoxy resin. The epoxy resin composition preferably also comprises from 0.01 to 4 percent, more preferably from 0.05 to 2 percent of a curing catalyst, such as a 2-methylimidazole, based on the weight of the epoxy resin. The epoxy resin composition may comprise a cure inhibitor, such as boric acid, for modifying the curing catalyst. The amount of such cure inhibitor, if present, preferably is up to 4 percent, more preferably up to 2 percent, based on the weight of the epoxy resin.

In the epoxy resin composition the total weight of the components i) epoxy resins, ii) curing agents for an epoxy resin, iii) curing catalysts, and iv) cure inhibitors is generally from 35 to 95 percent, preferably from 45 to 75 percent, more preferably from 50 to 70 percent, based on the total weight of components i) to iv) and the solvent present in the epoxy resin composition.

The entire amount of the solvent in the epoxy resin composition may consist of the solvent composition of the present invention. However, the epoxy resin composition may also comprise other solvents, such as larger amounts of ketones than recommended for the solvent composition of the present invention. Generally from 25 to 100 percent, preferably from 40 to 100 percent, more preferably from 40 to 60 percent of the solvent present in the epoxy resin composition originates from the solvent composition of the present invention, the residual amount being a known solvent or diluent for epoxy resins, such as a ketone.

The epoxy resin compositions are useful for various known applications, for example for preparing electrical laminates, and coatings. At least the preferred embodiments of the epoxy resin composition are homogeneous and generally have a viscosity that is low enough to allow a good impregnation of a reinforcing material, such as glass roving or reinforcing mats to produce reinforced epoxy compositions which cure upon heating. Techniques of impregnating reinforcing materials with epoxy resin compositions and curing the epoxy resin compositions are well known in the art.

The composition comprising one or more compounds selected from i) epoxy resins, ii) curing agents for an epoxy resin, iii) curing catalysts, and iv) cure inhibitors in the solvent composition of the present invention may comprise further components in addition to those mentioned above. For example, the composition may comprise solid particles, such as a filler. A filler which increases the fire retardant properties of the composition is particularly useful. However, the composition does not comprise more than 50 percent, preferably not more than 30 percent, most preferably not more than 25 percent of solid particles, based on the total weight of the composition.

The term "comprising" as used herein means "including". The term "comprising" is not to be understood to mean "consisting of".

The present invention is further illustrated by the following examples which should not be construed to limit the scope of the present invention. All parts and percentages are by weight unless otherwise mentioned.

Examples 1 to 8 and Comparative Examples A to C

Solutions of dicyandiamide ("DICY") in various individual solvents and solvent compositions are prepared. The concentrations are listed in Table I below. The produced solutions are visually inspected at room temperature (RT) whether they are clear. Clear solutions are designated as "Y" (Yes). Those that are not clear are designated as "N" (No) in Table I below. The solutions that are

clear at room temperature are tested whether they remain clear after they have been stored at 5°C for 24 hours.

Furthermore, Table I lists whether the solutions contain harmful or toxic chemicals, as defined by the EU criteria, which is a Classification according to Annex I of Council Directive 67/548/EEC (Dangerous Substances Directive).

Table I

(Comparative) Example	A	B	C	1	2	3	4	5	6	7	8
DICY SOLUTION	Wt. Pts	Wt. Pts	Wt. Pts	Wt. Pts	Wt. Pts	Wt. Pts	Wt. Pts	Wt. Pts	Wt. Pts	Wt. Pts	Wt. Pts
DICY	3	3	3	3	3	3	3	3	3	3	3
Propylene glycol methyl ether	34.4	29.4	26.0	31	25.0	27.3	24.3	31	25	28.3	26.3
Water	--	5	4.7	--	4.5	2.5	5.4	4	4	2	4.5
Propylene Carbonate	--	--	--	10	4.5	4.5	5	5	5	--	--
Ethylene carbonate										5	4.5
Dimethyl formamide			4.6								
Properties											
Wt.-% DICY *	8.7	8.7	8.5	7.3	8.8	8.7	8.7	7.5	8.8	8.5	8.5
Clear DICY Soln RT	N	Y	Y	N	Y	Y	Y	Y	Y	Y	Y
Stable at 5°C for 24hrs	-	Y	Y	-	Y	Y	Y	Y	Y	Y	Y
Contains harmful or toxic chemicals	N	N	Y	N	N	N	N	N	N	N	N

* wt.-% DICY, based on total amount of solvents

The DICY (dicyandiamide) solutions in Examples 2 to 8 in Table 1 are clear and do not contain harmful or toxic chemicals as defined by the above-mentioned EU criteria. The same is true for Comparative Example B. However, as illustrated by Comparative Examples O and P in Table 4 below, a solvent composition comprising propylene glycol methyl ether and water without the presence of a carbonate is sensitive towards changes in the chosen epoxy resin. The comparison between Examples 1 and 5 illustrates that advantageously a third component c), such as water, is included in the solvent composition of the present invention if the solvent composition comprises propylene carbonate.

Examples 9 to 20 and Comparative Examples D to P

Solutions of dicyandiamide ("DICY") in various individual solvents and solvent compositions are prepared. The solutions are blended with an epoxy resin solution. In Examples 9 to 13 and Comparative Examples D to J the epoxy resin is dissolved in propylene glycol methyl ether or ethylene glycol methyl ether. In Examples 14 to 18 and Comparative Examples K and L the epoxy resin is dissolved in methyl ethyl ketone. In Examples 19 and 20 and Comparative Examples M to P the epoxy resin is dissolved in acetone.

With the exception of Example 20 and Comparative Example P a brominated epoxy resin in methyl ethyl ketone is used which is commercially available under the trademark D.E.R. 539 EK80 from The Dow Chemical Company. In Example 20 and Comparative Example P a brominated epoxy resin in acetone is used which is commercially available under the trademark D.E.R. 592 A80 from The Dow Chemical Company.

The compositions of the final blends are listed in Tables 2 to 4 below. If the produced blend is clear and homogeneous at room temperature, it is classified as being compatible with the epoxy resin and is designated as "Y". Otherwise, for example if the resulting blend is turbid or contains two phases, it is designated as "N".

Laminates are produced and visually inspected as to whether dicyandiamide crystals are visible and if yes, to what extent. The laminates are classified as unacceptable laminate quality ("U"), acceptable laminate quality ("ACC"), standard laminate quality ("ST"), and superior laminate quality)("SUP").

Furthermore, Tables 2 to 4 list whether the solutions contain harmful or toxic chemicals, as defined by the EU criteria, which is a Classification according to Annex I of Council Directive 67/548/EEC (Dangerous Substances Directive).

Table II

(Comparative) Example	D	E	F	G	H	I	J	9	10	11	12	13
Composition	Wt. Parts	Wt. Parts	Wt. Parts	Wt. Parts	Wt. Parts	Wt. Parts	Wt. Parts	Wt. Parts	Wt. Parts	Wt. Parts	Wt. Parts	Wt. Parts
Epoxy Resin	100	100	100	100	100	100	100	100	100	100	100	100
Propylene glycol methyl ether		49.3	53.3	56.7	58.7	39.6	58.1	63.4	60	63.6	75	73.3
Solid DICY	3	3	3	3	3	3	3	3	3	3	3	3
Propylene Carbonate								3.3	4.7			4.5
Ethylene Carbonate										5	5	
Ethylene glycol methyl ether	63.4											
Dimethyl formamide	3.3	10.7	6.7		4.7	4.6	4.6					
Water		6.7		10	4.4	4.7	4.7		2		2	4.5
Acetone							6.3					
2-Phenylimidazole (20%)	0.8						0.8				0.8	0.8
Properties												
Compatible with epoxy resin	Y	N	N	N	Y	Y	Y	N	Y	Y	Y	Y
Laminate Quality	St	-	-	-	-	-	St	-	-	-	St	Sup
Contains harmful or toxic chemicals	Y	Y	Y	N	Y	Y	Y	N	N	N	N	N

" - " means: not tested

Table III

(Comparative) Example	14	15	16	17	18	K	L
Composition	Wt. Parts	Wt. Parts	Wt. Parts	Wt. Parts	Wt. Parts	Wt. Parts	Wt. Parts
Epoxy Resin	100	100	100	100	100	100	100
Methyl ethyl ketone	25	25	25	25	25	34.3	25
Propylene glycol methyl ether	25	30	31	30	25	26.4	18.5
Solid DICY	3	3	3	3	3	3	3
Propylene carbonate	4	9	5	3	3	--	--
Dimethyl formamide							13.5
Water	5	1	4	7	7	8	
2-Phenylimidazole (20%)			0.8				0.8
Properties							
Compatible with epoxy resin	Y	Y	Y	N	N	N	Y
Laminate Quality	-	-	STD	-	-	-	STD
Contains harmful or toxic chemicals	N	N	N	N	N	N	Y

" - " means: not tested

Table IV

(Comparative) Example	M	N	O	P	19	20
Composition	Wt. Parts	Wt. Parts	Wt. Parts	Wt. Parts	Wt. Parts	Wt. Parts
Epoxy Resin	100	100	100	100*	100	100*
Acetone	34.3	34.3	25	25	25	25
Propylene glycol methyl ether	27.9	29.4	35.9	36	30.9	30.9
Solid DICY	3	3	3	3	3	3
Propylene carbonate					5	5
Water	6.5	5	4	4	4	4
2-Phenylimidazole (20%)			0.8	0.8	0.8	0.8
Properties						
Compatible with epoxy resin	N	Y	Y	N	Y	Y
Laminate Quality	-	ACC	ST	-	SUP	-
Contains harmful or toxic chemicals						

" - " means: not tested

*Contains D.E.R. 592 A80 (Trademark) instead of D.E.R. 539 EK80 (Trademark)

The compositions of Examples 10 to 13, 19 and 20 are all compatible with the epoxy resin, that means that these formulations are clear and homogeneous, even in the absence of dimethyl

formamide, which is classified as harmful or toxic by the above-mentioned EU criteria.

The comparison between Examples 9 and 10 illustrates that advantageously a third component c), such as water, is included in the solvent composition of the present invention if the solvent composition contains propylene carbonate. Examples 11 and 12 illustrate that the compositions are compatible with the epoxy resin with or without a third component c), such as water, if the solvent composition contains ethylene carbonate.

Examples 14 to 18 illustrate that it is advisable to keep the water content within the more and most preferred range, that means from 2 to 15 percent, most preferably from 5 to 12 percent, based on the total weight of propylene glycol methyl ether (component a), propylene carbonate (component b), and water (component c), if the epoxy resin is dissolved in methyl ethyl ketone. The compositions of Comparative Examples D-M and P are either not compatible with the epoxy resin or contain dimethyl formamide, which is classified as harmful or toxic by the above-mentioned EU criteria.

The composition of Comparative Examples O and P is sensitive towards changes in the chosen epoxy resin. While the composition of Comparative Example O is compatible with the D.E.R. 539 EK80 (Trademark) epoxy resin, the corresponding composition of Comparative Example P is not compatible with the D.E.R. 592 A80 epoxy resin. The comparison between Examples 19 and 20 illustrates that the solvent composition of the present invention, specifically the solvent composition comprising propylene glycol methyl ether, propylene carbonate and water, is compatible with two different types of epoxy resins.

CLAIMS:

1. A solvent composition comprising

a) from 60 to 90 percent of a glycol monoether,

b) from 5 to 15 percent of an organic low molecular weight compound containing a carbonate group and

c) from 0 to 25 percent of a protic liquid other than a glycol monoether or diether,

based on the total weight of a), b) and c).

2. The solvent composition of Claim 1 wherein component a) is a propylene or butylene glycol monoether represented by Formula I

$$R^1O - (CH_2 - CHR^3O)_n - R^2 \qquad (I)$$

wherein

one of the substituents R^1 and R^2 is an alkyl group having from 1 to 12 carbon atoms and the other of the substituents R^1 and R^2 is hydrogen,

R^3 in each occurrence independently is methyl or ethyl and

n is from 1 to 4.

3. The solvent composition of Claim 2 wherein in Formula I one of the substituents R^1 and R^2 is an alkyl group having from 1 to 4 carbon atoms and the other of the substituents R^1 and R^2 is hydrogen, R^3 in each occurrence is methyl and n is 1, 2 or 3.

4. The solvent composition of any one of Claims 1 to 3 wherein component b) is butylene carbonate, propylene carbonate or ethylene carbonate.

5. The solvent composition of any one of Claims 1 to 4 wherein component c) is water.

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6. The solvent composition of any one of Claims 1 to 5 comprising a) a propylene glycol monoether, b) propylene carbonate and c) water.

7. The solvent composition of any one of Claims 1 to 5 comprising a) a propylene glycol monoether and b) ethylene carbonate.

8. A composition comprising one or more compounds selected from

i) epoxy resins,

ii) dicyandiamides,

iii) curing catalysts selected from tertiary amines and heterocyclic amines, and

iv) cure inhibitors selected from boric acid, metaboric acid, boric acid anhydride, maleic acid and mixtures of boric acid or metaboric acid with fluoroboric acid,

in a solvent composition comprising a) a glycol monoether or diether and b) an organic low molecular weight compound containing a carbonate group.

9. The composition of Claim 8 wherein the solvent composition comprises a) at least 45 percent of a glycol monoether or diether, b) from 1 to 30 percent of an organic low molecular weight compound containing a carbonate group and c) from 0 to 25 percent of a protic liquid other than a glycol monoether or diether, based on the total weight of a), b) and c).

10. The composition of Claim 8 comprising a solvent composition set forth in any one of claims 1 to 7.

11. A method of preparing a composition comprising one or more compounds selected from

i) epoxy resins,

ii) dicyandiamides,

iii) curing catalysts selected from tertiary amines and heterocyclic amines, and

iv) cure inhibitors selected from boric acid, metaboric acid, boric acid anhydride, maleic acid and mixtures of boric acid or metaboric acid with fluoroboric acid,

in a solvent composition by contacting one or more such compounds with

a) a glycol monoether or diether and

b) an organic low molecular weight compound containing a carbonate group.

12. Use of

a) a glycol monoether or diether and

b) an organic low molecular weight compound containing a carbonate group

as a diluent for one or more compounds selected from

i) epoxy resins,

ii) dicyandiamides,

iii) curing catalysts containing a tertiary amine group or a heterocyclic amine group, and

iv) cure inhibitors selected from boric acid, metaboric acid, boric acid anhydride, maleic acid and mixtures of boric acid or metaboric acid with fluoroboric acid.

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(54) **Title:** SOLVENT COMPOSITION

(57) **Abstract:** A solvent composition which comprises a) a glycol monoether or diether, b) a protic liquid other than a glycol monoether or diether; and c) an organic liquid containing an amide or carboxyl group, is particularly useful as a diluent for epoxy resins, curing agents for an epoxy resin, curing catalysts, and/or cure inhibitors.

DECLARATION AND POWER OF ATTORNEY

USA/PCT

As a below named inventor, I hereby declare that:

(a) My residence and Citizenship are as stated below my name My P.O (mailing) address is the same as my residence unless otherwise stated

(b) I verily believe I am/we are the original, first and sole/joint inventor(s) of the subject matter that is embraced by and for which a patent is sought on the invention entitled: **SOLVENT COMPOSITION**

and the specification of which (check one) ☐ is attached hereto (_____)
☒ was filed on September 8, 2000 as (44599).
Application No. PCT/US00/24915
and was amended on _____

(c) I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment referred to above.

(d) I acknowledge my duty under 37 CFR 1.56 to disclose to the U.S. Patent and Trademark Office all information known to me to be material to patentability as defined in 37 CFR 1.56(b). If this application is a continuation-in-part application, I acknowledge the duty to disclose all information known to me to be material to patentability as defined in 37 CFR 1.56(b) that became available between the filing date of the prior application from which priority is claimed in part (f) below, and the national or PCT international filing date of this application

(e) I hereby claim foreign priority benefits under 35 U.S.C. § 119(a)-(d) or §365(b) of any foreign application(s) for patent or inventor's certificate listed below or §365(a) of any PCT international application that designated at least one country other than the United States of America listed below, and also identify below any other foreign equivalent application for patent or inventor's certificate or any other equivalent PCT international application having a filing date before that of the application on which priority is claimed:

PRIOR FOREIGN APPLICATION(S) Number	Country or PCT	Day/Month/Year Filed	PRIORITY CLAIMED	CERTIFIED COPIES INCL.
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☐ Additional claims for benefit are attached.

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US or PCT Appln. Serial No.	Filing Date	Status at Application Filing Date
60/155,825	September 23, 1999	Pending

☐ Additional claims for benefit are attached.

I hereby appoint the attorney(s) and/or agent(s) at the following Customer No to prosecute this application and to transact all business in the Patent and Trademark Office connected therewith. Address all correspondence to The Dow Chemical Company's appointed counsel at:

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This appointment, including the right to delegate this appointment, shall also apply to the same extent it is applicable under the laws of the United States of America to any proceedings established by the Patent Cooperation Treaty

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements are made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both under 18 U.S.C. § 1001 and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

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